Fermi surface and low temperature structure in (DMET-TSeF)$_2$Au(CN)$_2$

K. Kato$^{a,*}$, K. Oshima$^b$, T. Kambe$^b$, Y. Nogami$^b$, T. Sasaki$^c$, M. Motokawa$^c$, R. Kato$^d$

$^a$IMS, Okazaki 444-8585, Japan
$^b$GNST, Okayama University, Okayama 700-8530, Japan
$^c$IMR, Tohoku University, Sendai 980-8577, Japan
$^d$RIKEN, Wako, 351-0198, Japan

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Abstract

The title compound is one of the DMET-TSeF salts which are quasi-one-dimensional organic metals at room temperatures. We show that the compound has a closed Fermi surface using the angle-dependent magnetoresistance oscillation (AMRO) method together with magnetoresistance and magnetization quantum oscillation measurements at low temperatures. By the X-ray measurement, we confirm that the structural phase transition observed around 180 K is related to the anomaly observed in the temperature dependence of the resistivity [1]. The nesting vector has the direction of 0.5$a^*+0.5c^*$. The obtained low temperature closed Fermi surface has about 4% of the first Brillouin zone area. The AMRO data show the existence of a very anisotropic Fermi surface where the diameter perpendicular to the most conducting $a$ direction is about 0.5 $c^*$. The possible origin of the low temperature closed Fermi surface is discussed and the relation to the ordering of anion molecules is considered as the probable reason.

Keywords: Transport Measurement, X-ray diffraction, Organic conductors

1. Introduction

DMET-TSeF is a hybrid molecule made from a half of the TMTSF and a half of the BETS molecules. The DMET-TSeF salts have quasi-one-dimensional Fermi surfaces at room temperature, and most of them show remarkable physical properties such as FISDW, superconductivity and so on. The low temperature Fermi surfaces are considered to be also quasi-one-dimensional, though the extent of the warping changes. (DMET-TSeF)$_2$Au(CN)$_2$ is also a quasi-one-dimensional organic conductor in the same family, and it is metallic down to the lowest temperature so far measured. But the transport and magnetization measurements show the existence of a closed Fermi surface at low temperatures [2]. It has been noticed that the clear anomaly exists around 180 K in the temperature dependence of the resistivity [1], therefore a possible structural transformation has been expected. But the low temperature structural investigation has been unsuccessful due to the fragility of the crystals. The present paper reports for the first time on the successful low temperature structural measurements. We discuss the relation between the phase transition and the possible Fermi surface in conjunction with the recently obtained AMRO type transport measurements at low temperatures.

Fig. 1. AMRO in the plane perpendicular to $a$ direction at 24 T. Integers corresponding to the peak positions are shown.

* Corresponding author. Tel: +81-564-55-7470; fax: +81-564-55-7448; E-mail: kkato@imr.ac.jp
2. Results and Discussion

The samples measured have been obtained using standard electrochemical oxidation of the DMET-TSeF molecule [1]. The low temperature crystal structure has been studied around 100 K, using the X-ray structural analysis system (R-AXIS, Rigaku) at the VBL of Okayama University. The Fermi surface has been studied by AMRO method [3] using the hybrid magnet of the Tohoku University. In the AMRO measurement, the peaks obey the simple formula indicated in Fig. 1, where the angle $\theta$ has been measured from the normal direction to the two-dimensional conduction plane. The data in Fig. 1 were obtained at 4.2 K, therefore the signal due to the quantum oscillation is not large except around $n=1$ peak. The depression of the AMRO peak at $n=1$ is seen, but the reason is not clear at present. The magnetoresistance shows very sharp angular oscillation for $n>1$, and the result gives the diameter of the Fermi surface along $c^*$ as 0.5$c^*$. By the de Haas–van Alphen effect and Shubnikov–de Haas effect, we observe the quantum oscillation with the period of 0.003T$^{-1}$ when the magnetic field is applied along $b^*$. We find that the area of the Fermi surface is about 4 % of the first Brillouin zone, therefore the diameter along $a^*$ direction should be less than 0.1$a^*$.

![Fig. 2. The X-ray photograph at 92.5 K. New spots corresponding to the superstructure appear among the indexed Bragg peaks.](image)

From the X-ray structural measurements, we found a superstructure with the nesting vector of $0.5a^* + 0.5c^*$ (Fig. 2). The new spots among the indexed Bragg peaks appear below 180 K. The result clearly shows the existence of commensurate modulation to the lattice. The temperature dependence of the reflection intensity corresponding to the superstructure is shown in Fig. 3. It is clear that the structural phase transition occurs around 180 K and the temperature corresponds to the anomaly in the temperature dependence of resistivity as noted above. The low temperature structure is not yet completely obtained, but it seems that the phase transition is related to the anion structure. As can be seen from the room temperature structural analysis, the thermal motion of CN atoms are extraordinarily large, therefore the phase transition can be related to the structural order of the Au(CN)$_2$ anions. Preliminary measurements suggest a new anion ordered structure along $b^*$ related to the bending of the Au(CN)$_2$ molecules, but further experimental studies are necessary to get conclusive results.

![Fig. 3. The temperature dependence of the reflection intensity corresponding to the superstructure.](image)

The possible Fermi surface reconstructed from the room temperature one [1] due to the nesting vector is schematically shown in Fig. 4. The size of the newly obtained closed Fermi surface is nearly consistent with the result obtained by the AMRO measurement. We consider that the origin of the phase transition at 180 K is not electronic. The reason is that the best nesting direction from the room temperature Fermi surface is not $0.5a^* + 0.5c^*$ but $0.5a^*$. Therefore, we consider that the transition is produced to stabilize the structure in the anion layer and the packing of the donor molecules through the contraction of lattice parameters by cooling. The feasibility of the model can be confirmed if we perform the low temperature structural analysis.

In conclusion, we found that the low temperature Fermi surface of (DMET-TSeF)$_2$Au(CN)$_2$ is very anisotropic by the AMRO measurement. The use of the high field hybrid magnet is very effective to study small Fermi surfaces because we need large angle data where satisfying the condition cot $\theta > 1$ becomes difficult.

![Fig. 4. The schematic reconstruction of the Fermi surface due to the nesting vector $0.5 a^* + 0.5 c^*$ shown by an arrow.](image)

References